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70 YEARS OF CREATING TOMORROW



**Los Alamos**  
NATIONAL LABORATORY

## **Keeping the Momentum and Nuclear Forensics at Los Alamos National Laboratory**

Robert Steiner, Heather Dion, Donald Dry, William Kinman,  
Stephen LaMont, David Podlesak and Lav Tandon



# Keeping the Momentum

Following the conclusion of the 2016 Nuclear Security Summit 5 organizations were entrusted with ensuring the legacy created by the NSS process:

- IAEA
- GICNT
- INTERPOL
- Global Partnership

# Nuclear Forensic Science Fundamental Requirement: Is it ours?



- If nuclear material is found outside of administrative controls anywhere in the world, then each country should be able to answer the question:

“Is this consistent with our material?”

- States have a responsibility to establish a system for identifying if materials found out of regulatory control are consistent with those used, produced, or stored within their borders
- A national nuclear forensic library is extremely valuable for answering this question with timeliness and confidence





# GICNT: Building Global Nuclear Forensics Awareness

## Multilateral and regional awareness raising efforts

- **IAEA Nuclear Security Series**
  - NSS #2-G Nuclear Forensics in Support of Investigations
  - NST018: Development of a National Nuclear Forensics Library
  - NST015: Analytic Measurements for Materials out of Regulatory Control in Support of a Nuclear Forensics Investigation
- **IAEA Training**
  - Introduction to Nuclear Forensics
  - Nuclear Forensics Methodologies
- **ASEAN Regional Forum engagement**





# Global Initiative to Combat Nuclear Terrorism

## Integrate

Integrate collective experience and opportunities to strengthen the overall global architecture to combat nuclear terrorism

## Experience

Bring together experience and expertise from the nonproliferation, counterproliferation and counterterrorism disciplines

## Opportunity

Provide opportunities for partner nations to share information and expertise in a voluntary, non-binding framework

Implementation and Assessment Group

Nuclear Detection Working Group

Nuclear Response and Mitigation Working Group

Nuclear Forensics Working Group



# LANL Participation in NFWG Activities



LANL has supported the NFWG Chair (D.Hill and T.Bull since the inception of the IAG)

- Support to Plenary and IAG Meetings
- Documents
  - Fundamentals Document
  - Information Sharing
  - Exercise Playbook
- Exercises
  - Iron Koala (2012)
  - Blue Beagle (2014)
  - Tiger Reef (2014)
  - Mystic Deer (2015)
  - Radiant City (2015)
  - Maple Sunrise (2015)
  - Northern Lights (2015)
  - Kangaroo Harbor (2016)



GICNT

**IRON KOALA:** Information Sharing During Nuclear Smuggling Events  
SYDNEY, AUSTRALIA | May 28-30, 2012





# Bilateral Projects: Expand the understanding of nuclear attribution and signatures

## Bilateral projects of interest to the international nuclear forensics community

### Partners Engaged

- China
- France
- Japan
- EURATOM
- Canada
- Australia
- South Korea

Radiochronometers provide insight into separation date and quality

### Areas for improvement

- Certified Reference Materials
  - Development of  $^{229}\text{Th}$
- Measurement and interpretation of multiple parent-progeny pairs
  - $^{230}\text{Th}/^{234}\text{U}$  and  $^{231}\text{Pa}/^{235}\text{U}$

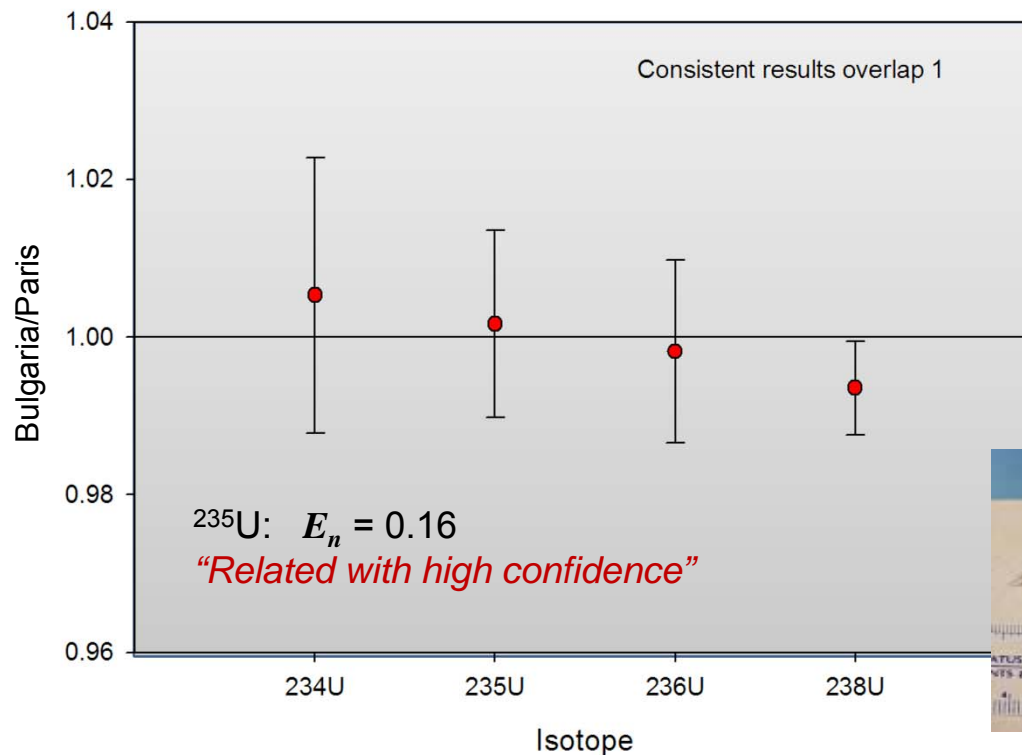
### Current Focus: Uranium Chronometry

- Work with partners to increase precision and accuracy in uranium age dating
- Focus on a single topic to maximize INFC's investment



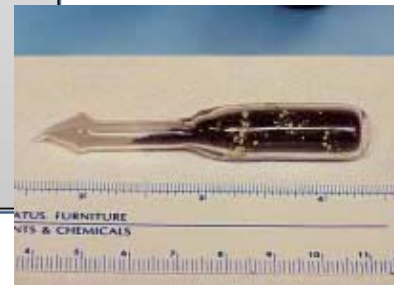
# 1999 Bulgaria and 2001 Paris Comparison

## Comparison of Bulgaria and Paris Uranium Isotopic Composition Results



### Demonstrated:

- Connections between cases
- Value of libraries
- Importance of international cooperation



Bulgaria sample results from: *Forensic Analysis of a Smuggled HEU Sample Interdicted in Bulgaria*, Lawrence Livermore National Laboratory, U.S. Dept. of Energy, UCRL-ID-143216, 2001.

France sample results from the proceedings of: *International Conference on Illicit Nuclear Trafficking: Collective Experience and the Way Forward*, IAEA-CN-154/062, November, 2007.

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# Nuclear Forensics Partner Organizations

- United States
  - Lawrence Livermore National Laboratory
  - Los Alamos National Laboratory
  - Pacific Northwest National Laboratory
  - Argonne National Laboratory
  - Department of State
  - Department of Homeland Security
  - Federal Bureau of Investigation
- Algeria
  - Commissariat à l'Energie Atomique
- Australia
  - Australia Nuclear Science and Technology Organization
- Canada
  - Canada Nuclear Safety Commission
- China
  - China Institute for Atomic Energy
- European Commission
  - Joint Research Center-Institute for Transuranium Elements
- France
  - Commissariat à l'Energie Atomique
- International Atomic Energy Agency
  - Division of Nuclear Security
- Japan
  - Japan Atomic Energy Agency
- Kazakhstan
  - Kazakhstan Atomic Energy Commission
- Republic of Korea
  - Korea Atomic Energy Research Institute
  - Korea Institute of Nuclear Nonproliferation and Control
- Russia
  - ROSATOM
- South Africa
  - Nuclear Energy Corporation of South Africa



# How does LANL Provide Sound Technical Support to the International Community?

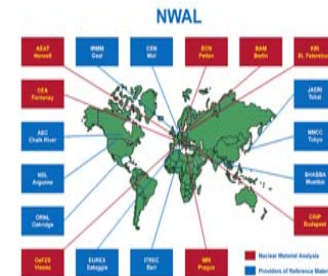
*Our ability to characterize nuclear materials and processes relies on analytical methods from:*

Certification of materials in pit production



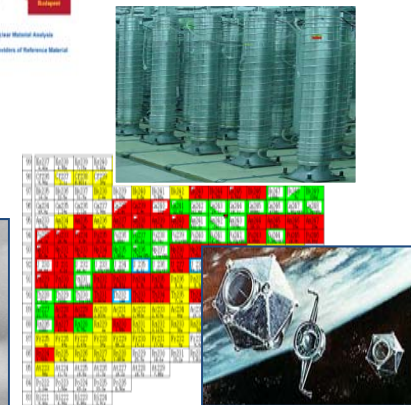
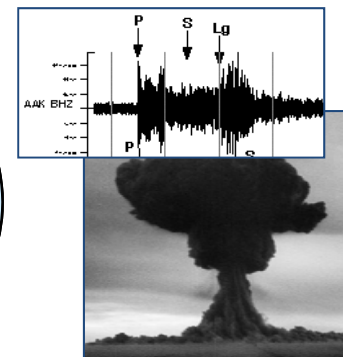
*Our ability to characterize signatures of worldwide nuclear materials production relies on skills built in:*

Support for international safeguards programs



*Our ability to characterize the origins of a nuclear explosion is based on:*

Underground test experience







# Facilities to Work with Materials of all Quantities

All facilities house ongoing missions that exercise analytical capabilities routinely

TA-48



*Sigma*



CMR

TA-55



RC-45  $< 10^{11}$  atoms Pu-239



RC-1, Sigma



$10^9 - 10^{22}$  atoms Pu-239

Nuclear facilities (CMR, PF-4)



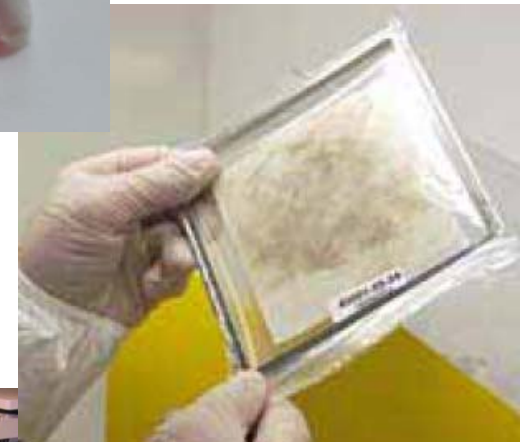
$> 10^{22}$  atoms Pu-239





## Sample Types

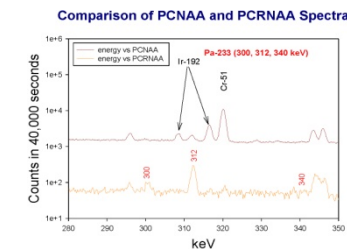
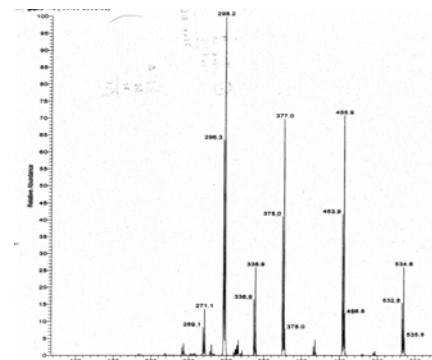
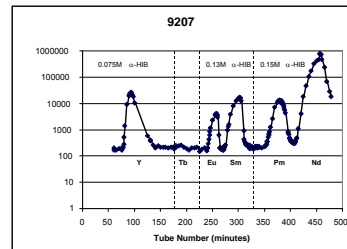
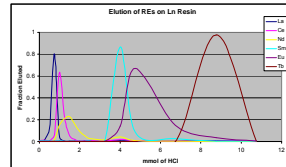
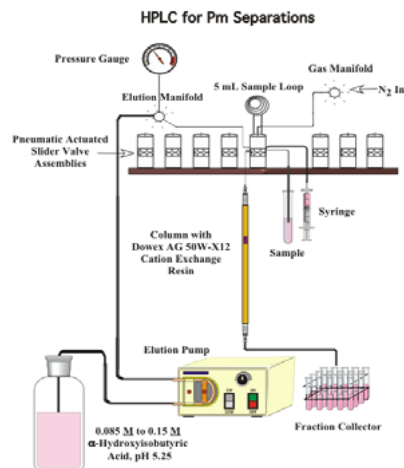
- Swipes
- Soil
- Water
- Vegetation
- Biological (urine, bone, tissue)
- Geological
- Radiological Materials





# Radiochemistry

- Single or multi-element
- Elementally separated radiochemistry
- Standardized procedures
- Refractory matrices





# Bulk Analysis

## Multi-collector ICP-MS

(MC-ICP-MS)

High precision, high accuracy

Isotope ratios (U, Th, Sr, Pb, Fe, B...)

ng to <fg sample requirements



## Sector Field ICP-MS

(SF-ICP-MS)

Ppq – ppm elemental concentrations



## Multi-collector TIMS

Pu, other actinides, Sr, Nd







# Summary

- LANL has 70 years of experience in nuclear forensics and supports the community through a wide variety of efforts and leveraged capabilities
  - Expanding the understanding of nuclear forensics
  - Providing training on nuclear forensics methods
  - Developing bilateral relationships to expand our understanding of nuclear forensic science
  
- LANL remains highly supportive of several key organizations tasked with carrying forth the Nuclear Security Summit messages
  - IAEA
  - GICNT
  - INTERPOL



# Back up slides





# Analysis and Characterization of Actinide Materials

## *Onsite Analytical Chemistry and Sample Management*



Coordinate sample receiving, shipping, and distribution at TA-55 and CMR

Onsite radiochemical and trace analysis

## *Assay and Classical Chemistry*



Coulometric titration  
Ceric titration  
Pu (III) and Pu (IV)  
U Assay by Davies Gray  
Fe and Si determination  
Loss on Ignition (LOI)  
Free acid determination  
Standard preparation

## *Plasma Spectroscopy*

Inductively Coupled Plasma-Mass Spectrometry  
Inductively Coupled Plasma- Atomic Emission Spectrometry



DC Arc Emission

Cold-Vapor Atomic Fluorescence

## *X-Ray Fluorescence (XRF) and X-Ray Diffraction (XRD)*



Fingerprint Detection Technology



# Analysis and Characterization of Actinide Materials

## Mass Spectrometry



High-Precision  
Gas Mass Spectrometry

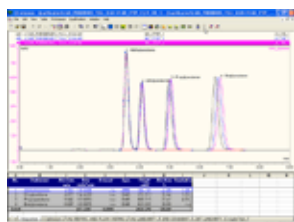


## Radiochemistry and Nondestructive Analysis

Alpha and gamma spectrometry  
Gross alpha, liquid scintillation



## Interstitial Analysis & Ion Chromatography



Fluoride, chloride, nitrite  
nitrate, phosphate, sulfate  
oxalate and perchlorate

carbon, oxygen, hydrogen  
sulfur, moisture, and tritium



## Laboratory Information Management System and Quality Assurance

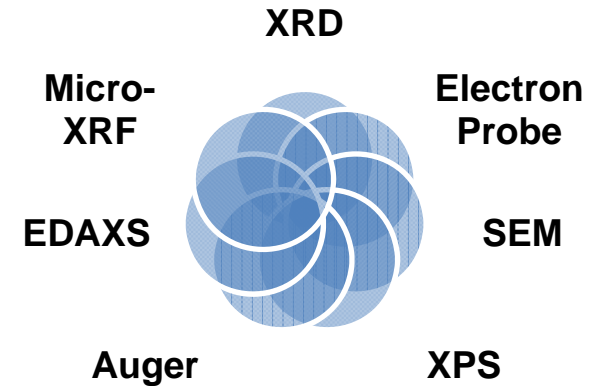
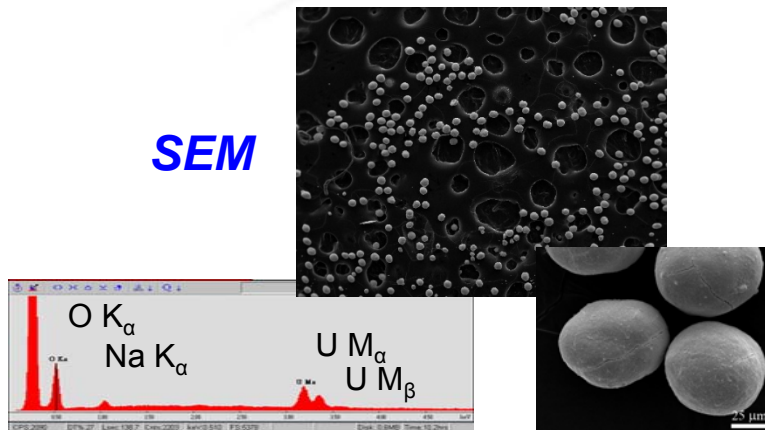
Oracle SQL\*LIMS  
Sample/nuclear material  
tracking and data  
management



Quality Assurance and Control  
Record Management  
Document Control  
Training

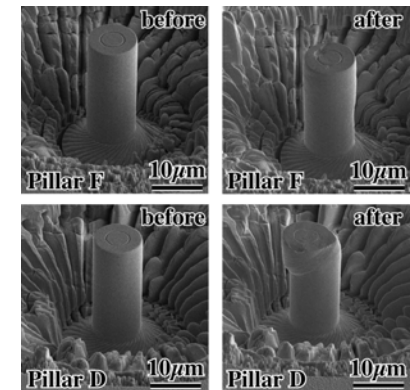
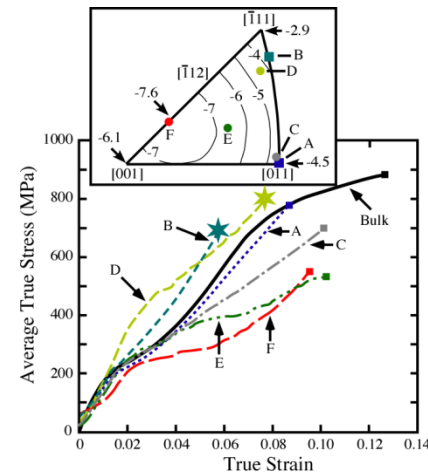
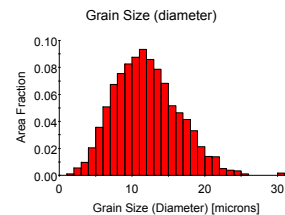
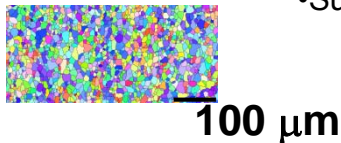


# Physical Characterization Tools for Bulk Materials



Materials processing techniques leave unique signatures in microstructures:

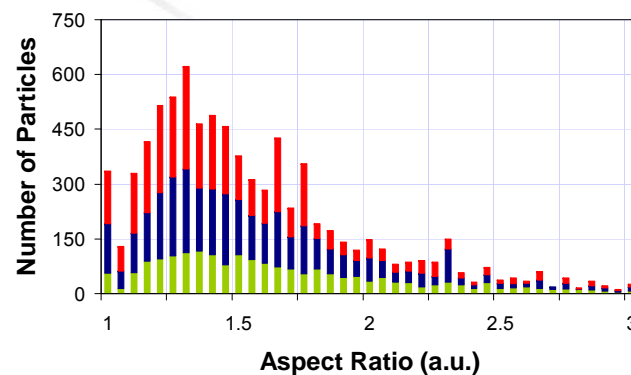
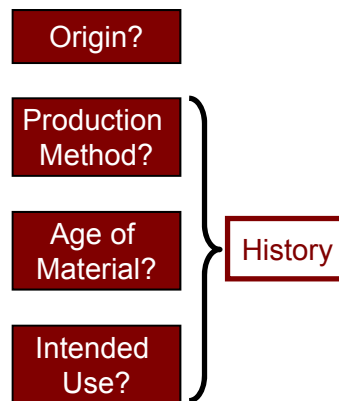
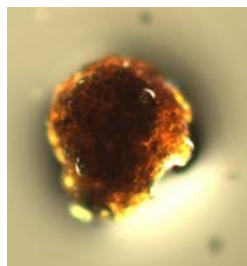
- Grain Size/Morphology
- Inclusion Distribution/Morphology
- Microstructural Texture
- Mechanical Properties
- Surface Features



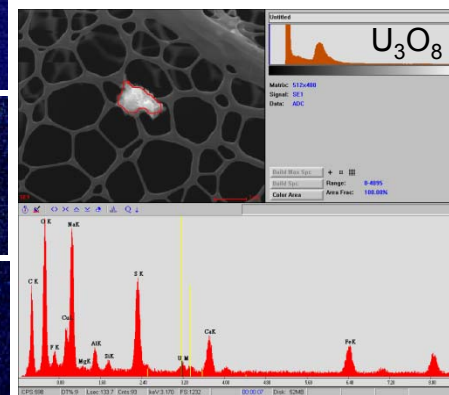
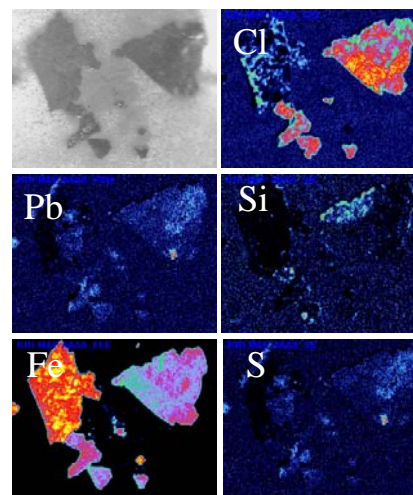
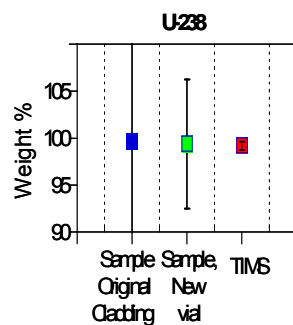




# Materials Analysis



Nuclide	Sample (μg/g)
$^{234}\text{U}$	$5.5 \times 10^1$
$^{235}\text{U}$	$5.7 \times 10^3$
$^{238}\text{U}$	$7.9 \times 10^5$
$^{238}\text{Pu}$	$7.4 \times 10^{-4}$
$^{239}\text{Pu}$	$2.5 \times 10^1$
$^{240}\text{Pu}$	$1.0 \times 10^{-2}$
$^{242}\text{Pu}$	$2.5 \times 10^{-4}$

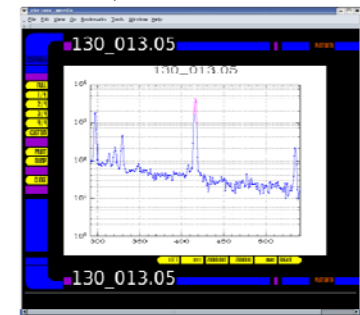




# Count Room Capabilities

Provides qualitative and quantitative assay of gamma, beta, and alpha-emitting radionuclides in a variety of matrices and over a wide range of activity levels,

- Trace levels to  $>10^{13}$  fissions
- Specializing in fission product measurements,
- Operates 24x7x365
- Sample receipt and handling protocols
- Makes ~ 70,000 measurements annually
  - 30,000 high resolution gamma collections
- 30 non-automated high-resolution gamma-ray spectrometers, some highly specialized
- 10 custom automated high-resolution gamma-ray spectrometers,
- 6 custom automated beta counters.
- 6 custom non-automated beta counters (3 ~0.2 CPM Bkg)
- ~ 90 Alpha spectrometers,
- Batch and interactive analysis codes,
- Relational database with web-based visualization tools.







# RC-45 Clean Chemistry and Mass Spectrometry Facility



- Annually certified clean laboratories
- Perchloric acid capability
- Ultra low-level sample handling
- Co-located clean radiochemistry and analysis capability
- Isotopic and elemental analysis



# Clean Sample Preparation

## Cleanroom compatible sample ashing

Class 100, low insulation  
ashing equipment for  
particle control



## Radiochemistry processing

Class 10-100, glassware cleaning,  
sample dissolution and digestion, ion  
exchange chemistry, sample dry  
down



## Sample loading areas

Class 10 -100,  
electroplating,  
carborizing, direct loading





## New Additions

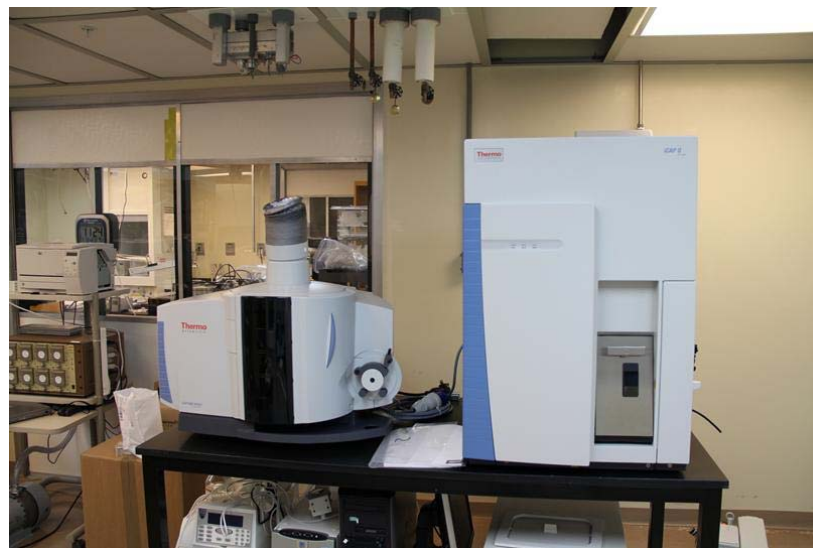


### **NuWave Micromill**

Small scale spatial sampling for  
Analysis by TIMS or ICP-MS

### **Quad ICP-MS and ICP-OES**

Trace analysis and coupling to  
on-line separations techniques  
and laser ablation for method  
development

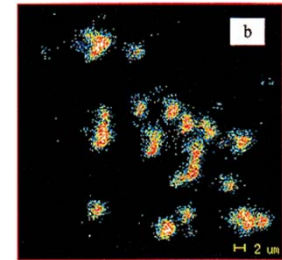




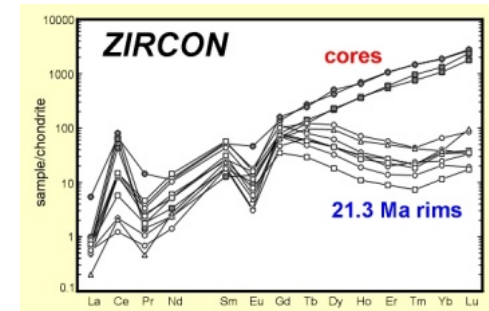


# Investments in Modern Capabilities

- Neptune MC-ICP-MS
- IsotopX Isoprobe-T MC-TIMS
- FEI SEM w/WDS & EDS
- Element XR ICP-MS



- Dual-Clover high resolution gamma-ray spectrometers





## Instruments Available

- Multi- Collector TIMS, 5 - IsotopX Phoenix instruments, 1 VG Sector 54
- Multi-Collector ICPMS - 2 Thermo Neptune Pluses, 1 Thermo Neptune, 1 Nu Instruments Plasma II.
- HR ICPMS - 2 Thermo Elements
- Quad ICPMS - 1 Thermo ICapQ, 1 PE Elan DRC, 1 Agilent 7700
- ICP-OES - Thermo ICap 6500
- XRF - Bruker S8 Lion
- SEM - FEI Field Emission Instrument
- Capillary electrophoresis and HPLC
- Several laser ablation systems
- Stable Isotopes - Thermo MAT 253 and Thermo Delta V
- Laser spectroscopy instrumentation





## Summary

- Analytical chemistry measurements on plutonium and uranium matrices are critical to numerous programs including safeguards accountancy verification measurements.
- Los Alamos National Laboratory operates capable actinide analytical chemistry and material science laboratories suitable for nuclear material and environmental forensic characterization.
- Los Alamos National Laboratory uses numerous means to validate and independently verify that measurement data quality objectives are met.
- Numerous LANL nuclear facilities support the nuclear material handling, preparation, and analysis capabilities necessary to evaluate samples containing nearly any mass of an actinide (attogram to kilogram levels).